

Vacuum-Induced Coherence in Ultracold Photoassociative Ro-Vibrational Excitations

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We show that coherence between two excited ro-vibrational states belonging to the same molecular electronic configuration arises quite naturally due to their interaction with electromagnetic vacuum. For initial preparation of a molecule in the desired ro-vibrational states, we propose to employ the method of ultracold photoassociation. Spontaneous decay of the excited molecule then gives rise to vacuum induced coherence between the excited ro-vibrational states. We demonstrate theoretically an interesting interplay of effects due to vacuum induced coherence and photoassociation. We apply our theory to photoassociation of bosonic Ytterbium (¹⁷⁴Yb) atoms which appear to be a promising system for exploring such interplay. The effects discussed here can be important for controlling decoherence and dissipation in molecular systems.

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Quantum coherences and interference form the basis of many fascinating phenomena in atomic and molecular systems [1, 2]. Its applications in spectroscopy, metrology, photovoltaics and information sciences have been most remarkable. One of the famous examples of quantum interference is the Fano effect [3, 4] that arises from interference of two competing optical transition pathways involving discrete and continuum states. Another notable coherence phenomenon is the vacuum-induced coherence (VIC) which arises due to quantum interference between two pathways of spontaneous emissions [5]. It is known that under appropriate conditions, VIC can lead to population trapping in excited states [2, 5]. This can be utilized in manipulating environment induced relaxation processes in a wide variety of systems such as atoms, ions, molecules, quantum dots [5–12], and has also been found to be effective against decoherence in quantum information processing [13]. One of the key conditions for VIC to occur is the nonorthogonality of the dipole moments of two spontaneous transitions. For atomic systems nonorthogonality is a stringent condition to achieve. Possible realization of VIC for an excited atom interacting with an anisotropic vacuum [14–16] and utilizing the $j = 1/2 \rightarrow j = 1/2$ transition in ¹⁹⁸Hg⁺ and ¹³⁹Ba⁺ ions have been suggested [17, 18]. Recently, a proof-of-principle experiment verifying its presence has been performed in quantum dots [19]. In spite of these attempts, a clear signature of VIC in atomic systems is yet to be obtained.

In this Rapid Communication we show that unlike atoms or ions, VIC arises quite naturally in molecular systems. This can be attributed to the quantum interference of spontaneous emission pathways from two ro-vibrational levels of an excited molecular electronic state to a ground molecular state. In this case, the required nonorthogonality condition is satisfied naturally as the two excited states belong to the same

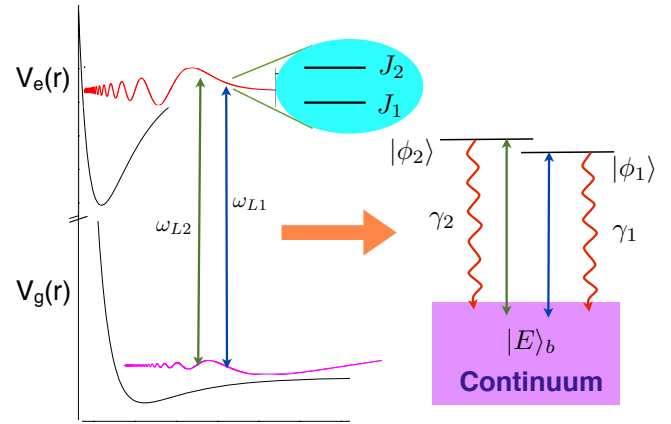


FIG. 1: (Color online) A schematic diagram for creation of coherence in two excited ro-vibrational levels by photoassociation. The two-atom continuum $|E\rangle_b$ is coupled to two ro-vibrational levels $|v, J_1\rangle(|\phi_1\rangle)$ and $|v, J_2\rangle(|\phi_2\rangle)$ of an excited molecular electronic state via two lasers of frequencies ω_{L1} and ω_{L2} , respectively. γ_1 and γ_2 are the spontaneous decay rates at which $|\phi_1\rangle$ and $|\phi_2\rangle$ decay to the continuum. Coherence arises between the two excited molecular states from the quantum interference of two spontaneous decay pathways $|\phi_1\rangle \rightarrow |E\rangle_b$ and $|\phi_2\rangle \rightarrow |E\rangle_b$.

molecular electronic configuration and differ only in rotational or vibrational quantum numbers. With the tremendous progress in high precision photoassociation (PA) spectroscopy [20, 21], low lying rotational levels can be selectively populated in a molecular excited state. This occurs due to PA transitions from the collisional continuum of two ground state cold atoms. VIC will be significant in such an atom-molecule interface system provided (i) there is no hyperfine interaction in the atoms that are photoassociated, (ii) there is no bound state close to the dissociation continuum of the ground molecular state and (iii) excited molecular levels have long life time.

However, to our knowledge, the possibility of VIC in such PA systems has not been addressed so far. As such, we propose here a novel PA scheme for realization of VIC in an

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atom-molecule system. We present results on the interplay of VIC and the effects induced by PA lasers such as dressing of the continuum. We show that the life time of the excited states can be controlled by appropriate manipulation of this interplay. Moreover, our results predict that for an optimum detuning and intensity of PA lasers, the interplay between VIC and PA can lead to coherent population trapping in the excited states.

The basic idea of our scheme is depicted in Fig. 1. We consider as our model a system of two excited molecular rovibrational levels $|v, J_1\rangle(|\phi_1\rangle)$ and $|v, J_2\rangle(|\phi_2\rangle)$ coupled to the two-atom ground continuum $|E\rangle_b$ via lasers. Initially either $|\phi_1\rangle$ or $|\phi_2\rangle$ is populated or partially both are populated via photoassociation of cold atoms using two lasers L_1 and L_2 of frequencies ω_{L1} and ω_{L2} , tuned near $|E\rangle_b \rightarrow |\phi_1\rangle$ and $|E\rangle_b \rightarrow |\phi_2\rangle$ transitions, respectively. Both the excited levels $|\phi_1\rangle$ and $|\phi_2\rangle$ decay spontaneously to the same ground continuum with decay rates γ_1 and γ_2 , respectively. The Hamiltonian governing the dynamics of this system can be written as $\mathcal{H} = \mathcal{H}_c + \mathcal{H}_{inc}$, where \mathcal{H}_c is the coherent part involving PA couplings and is given by,

$$\mathcal{H}_c = \sum_{n=1}^2 \hbar\omega_{bn} |\phi_n\rangle\langle\phi_n| + \int dE' |E'\rangle_b \langle E'| dE' + \int \sum_{n=1}^2 \left\{ \Lambda_{nE'} e^{-i\omega_{Ln}t} \hat{S}_{E'}^\dagger + \text{H.C.} \right\} dE'. \quad (1)$$

Here $\hbar\omega_{bn}$ are the binding energies of the bound states $|\phi_n\rangle$ ($n=1, 2$); $|E'\rangle_b$ is the bare continuum state and $\Lambda_{nE'} = \langle\phi_n| \vec{\mathcal{D}}_n \cdot \vec{E}_{Ln} |E'\rangle_b$ is the laser coupling for the transition from the n -th bound state to the bare continuum $|E'\rangle_b$. The vectors $\vec{\mathcal{D}}_n$ and \vec{E}_{Ln} are the dipole moment and electric field of the laser associated with the n -th transition, respectively. The operator $\hat{S}_{E'}^\dagger = |\phi_n\rangle_b \langle E'|$ is a raising operator. \mathcal{H}_c is exactly diagonalizable [22, 23] in the spirit of Fano's theory [3]. The eigen state of \mathcal{H}_c is the dressed continuum expressed as

$$|E\rangle_d = A_{2E}|\phi_2\rangle + A_{1E}|\phi_1\rangle + \int C_{E'}(E) |E'\rangle_b dE' \quad (2)$$

with the normalization condition $\langle E''|E\rangle = \delta(E - E'')$. Here $A_{nE} = \Lambda_{nE}(q\Gamma_{n'}/2 + \delta_{n'E})/\hbar[(\delta_{1E} + i\Gamma_1/2)(\delta_{2E} + i\Gamma_2/2) - \Gamma_1\Gamma_2(q - i)^2/4]$ with $n' \neq n$ and $C_{E'}(E) = \delta(E - E') + (A_{1E}\Lambda_{1E'} + A_{2E}\Lambda_{2E'})/(E - E')$. The term $\Gamma_n(E) = 2\pi|\Lambda_{nE}|^2/\hbar$, is the stimulated line width of the n -th bound state due to continuum-bound laser coupling and $\delta_{nE} = E/\hbar - \delta_n$ with detuning $\delta_n = \omega_{bn} - \omega_{Ln}$. Here $q = V_{12}/(\pi\Lambda_{1E}\Lambda_{2E})$ is analogous to the well-known Fano's q parameter [3] with $V_{12} = \mathcal{P} \int dE' \Lambda_{1E'}\Lambda_{2E'}/(E - E')$ where ' \mathcal{P} ' stands for principal value.

The incoherent part of the Hamiltonian \mathcal{H}_{inc} describes the interaction of vacuum field with the system and is given by,

$$H_{inc} = \int dE' \left[\sum_{n=1}^2 \sum_{\kappa,\sigma} g_{n,\sigma}(E', \kappa) \hat{S}_{E'}^\dagger \hat{a}_{\kappa,\sigma} e^{-i\omega_{\kappa}t} + \text{H.c.} \right] \quad (3)$$

where $\hat{a}_{\kappa,\sigma}$ is the annihilation operator of the vacuum field \vec{E}_{vac} and $g_{n,\sigma}(E', \kappa) = -\langle\phi_n| \vec{\mathcal{D}}_n \cdot \vec{E}_{vac}(\kappa) |E'\rangle_b$ is the dipole coupling with $\vec{E}_{vac}(\kappa) = (\sqrt{\hbar\omega_{\kappa}/2\epsilon_0 V}) \vec{\epsilon}_\sigma$, κ being the wave number, σ the polarization of the field and $\sqrt{\hbar\omega_{\kappa}/2\epsilon_0 V}$ the amplitude of the vacuum field. Let the joint state of the system-reservoir at a time t be expressed as,

$$|\Psi(t)\rangle = \sum_n a_n(t) |\phi_n, \{0\}\rangle + \int dE' \sum_{\kappa,\sigma} b_{E',\kappa,\sigma}(t) |E'\rangle_b |\{1_{\kappa,\sigma}\}\rangle \quad (4)$$

where a_n and $b_{E',\kappa,\sigma}$ are the amplitudes of n -th excited state and ground continuum, respectively. The state $|\phi_n, \{0\}\rangle$ corresponds to molecular excited state with field in vacuum and $|E'\rangle_b |\{1_{\kappa,\sigma}\}\rangle$ refers to (ground) bare continuum state with energy E' and one photon in mode κ of polarization σ . Using the standard Wigner-Weisskopf approach [2], after a long algebra we obtain,

$$\dot{\tilde{a}}_1 = (-\mathcal{G}_1 - i\delta_{12})\tilde{a}_1 - \mathcal{G}_{12}\tilde{a}_2 \quad (5)$$

$$\dot{\tilde{a}}_2 = -\mathcal{G}_2\tilde{a}_2 - \mathcal{G}_{21}\tilde{a}_1 \quad (6)$$

where \tilde{a}_n is the modified amplitude related to a_n by transformations, $\tilde{a}_1 = a_1 \exp[i(\tilde{\omega}_1 - \delta_{12})t]$ and $\tilde{a}_2 = a_2 \exp[i\tilde{\omega}_2 t]$. The dressed frequency $\tilde{\omega}_n$ and the detuning δ_{12} in the above expression are given by $\hbar\tilde{\omega}_n = \int |A_{nE}|^2 E dE$ and $\delta_{12} = (\omega_{L1} - \omega_{L2}) + (\tilde{\omega}_1 - \tilde{\omega}_2)$. The \mathcal{G}_n is the decay constant of the n -th bound state, given by

$$\mathcal{G}_n = \frac{1}{\hbar^2} \int dE' \int dt' \sum_{\kappa,\sigma} |g_{n\sigma}(E', \kappa)|^2 \exp \left[i \left(\omega_{Ln} + \tilde{\omega}_n - \frac{E'}{\hbar} - \omega_{\kappa} \right) (t - t') \right] \quad (7)$$

A key feature of Eqs. (5 – 6) is the coupling between the amplitudes via the cross damping term

$$\mathcal{G}_{nn'} \simeq \frac{1}{\hbar^2} \int dE' \int dt' \sum_{\kappa,\sigma} g_{n\sigma}(E', \kappa) g_{n'\sigma}^*(E', \kappa) \exp \left[i \left(\omega_{Ln} + \tilde{\omega}_n - \frac{E'}{\hbar} - \omega_{\kappa} \right) (t - t') \right] \quad (8)$$

where $n \neq n'$. For simplicity in writing the above equation we have assumed $(\omega_{L1} + \tilde{\omega}_1) \simeq (\omega_{L2} + \tilde{\omega}_2)$. It is important to understand that \mathcal{G}_{12} arises due to quantum interference of the spontaneous emission pathways resulting in VIC between the excited states amplitudes. Summing over the vacuum modes and then carrying out the time integral under Born Markov approximation, we finally obtain $\mathcal{G}_n = (3\pi\epsilon_0\hbar c^3)^{-1} \int dE' (\omega_{Ln} + \tilde{\omega}_n - E'/\hbar)^3 |\langle\phi_n| \vec{\mathcal{D}}_n |E'\rangle_b|^2$ and $\mathcal{G}_{12} = \mathcal{G}_{21} = (3\epsilon_0\hbar c^3\pi)^{-1} \int dE' \langle\phi_1| \vec{\mathcal{D}}_1 |E'\rangle \langle E'| \vec{\mathcal{D}}_2 |\phi_2\rangle (\omega_{L1} + \tilde{\omega}_1 - E'/\hbar)^{3/2} (\omega_{L2} + \tilde{\omega}_2 - E'/\hbar)^{3/2}$. Solving the coupled Eqs. (5) and (6) analytically, we obtain

$$\tilde{a}_1(t) = c_{1-} e^{z_- t} + c_{1+} e^{z_+ t} \quad (9)$$

$$\tilde{a}_2(t) = c_{2-} e^{z_- t} + c_{2+} e^{z_+ t} \quad (10)$$

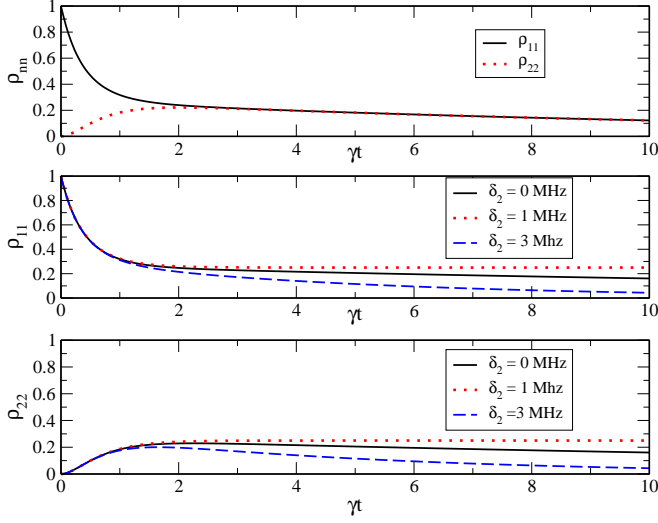


FIG. 2: (Color online) Uppermost Panel shows the plots of ρ_{11} and ρ_{22} in the absence of laser which is the case of normal VIC. Both ρ_{11} and ρ_{22} go to zero in the long time limit. In the middle and lower panel, the excited state population ρ_{11} and ρ_{22} are plotted as a function of dimensionless time γt for different values of the detuning of the second laser, δ_2 , keeping $\delta_1 = 0$. At $\delta_2 = 1$ MHz, the population is trapped between the two excited states. The intensities of L_1 and L_2 are 50 mW cm^{-2} and 0.1 mW cm^{-2} , respectively.

where, $z_{\pm} = \frac{1}{2}[-i\delta_{12} - \mathcal{G}_{\pm} \pm \Omega]$ and $c_{1\pm} = [\pm(-i\delta_{12} - \mathcal{G}_{\pm} \pm \Omega)\tilde{a}_1(0) \mp 2\mathcal{G}_{12}\tilde{a}_2(0)]/(2\Omega)$ and $c_{2\pm} = [\pm(i\delta_{12} + \mathcal{G}_{\pm} \pm \Omega)\tilde{a}_2(0) \mp 2\mathcal{G}_{12}\tilde{a}_1(0)]/(2\Omega)$ with $\Omega = \sqrt{(\mathcal{G}_{-} + i\delta_{12})^2 + 4\mathcal{G}_{12}^2}$, $\mathcal{G}_{-} = \mathcal{G}_1 - \mathcal{G}_2$ and $\mathcal{G}_{+} = \mathcal{G}_1 + \mathcal{G}_2$.

In the limit when laser intensities going to zero (weak coupling), we find $\tilde{\omega}_n \rightarrow (\omega_{bn} - \omega_{Ln})$, thus \mathcal{G}_n reduces to the usual damping constant γ_n . Moreover, for low energy we have

$$\mathcal{G}_{12} \simeq \gamma_{12} = \frac{\sqrt{(\omega_{b1}\omega_{b2})^3}}{(3\pi\epsilon_0\hbar c^3)} \langle \phi_1 | \vec{\mathcal{D}}_1 \cdot \vec{\mathcal{D}}_2 | \phi_2 \rangle. \quad (11)$$

Thus in the absence of lasers, the model reduces to normal VIC case in V-type system [2]. The above equation shows that γ_{12} vanishes if the molecular transition dipole moments $\vec{\mathcal{D}}_1$ and $\vec{\mathcal{D}}_2$ are orthogonal. In our model $\vec{\mathcal{D}}_1$ and $\vec{\mathcal{D}}_2$ are the transition dipole moments between the same ground and excited electronic states, therefore they are essentially nonorthogonal.

The excited state populations $\rho_{11} = |\tilde{a}_1|^2$, $\rho_{22} = |\tilde{a}_2|^2$ and the coherence $\rho_{12} = \tilde{a}_1\tilde{a}_2^*$ can be obtained from Eqs. (9) and (10). Explicitly

$$\begin{aligned} \rho_{nn}(t) = & \frac{e^{-2\mathcal{G}_1 t}}{4\Omega^2} [2\{A\Omega^2 - B\delta_{12}^2\}\rho_{nn}(0) \\ & - 8B\mathcal{G}_{12}^2\rho_{n'n'}(0) + 8B\mathcal{G}_{12}\delta_{12}\text{Im}[\rho_{12}(0)] \\ & - 8\mathcal{G}_{12}\Omega\text{Re}[\rho_{12}(0)]\sinh(\Omega t)], \end{aligned} \quad (12)$$

where $n' \neq n$, $A = [1 + \cosh(\Omega t)]$ and $B = [1 - \cosh(\Omega t)]$ and we have considered $\mathcal{G}_1 = \mathcal{G}_2$. At $4\mathcal{G}_1^2 = [4\mathcal{G}_{12}^2 - \delta_{12}^2]$, it follows from above equation that ρ_{11} and ρ_{22} become time-independent in long time limit meaning coherent population

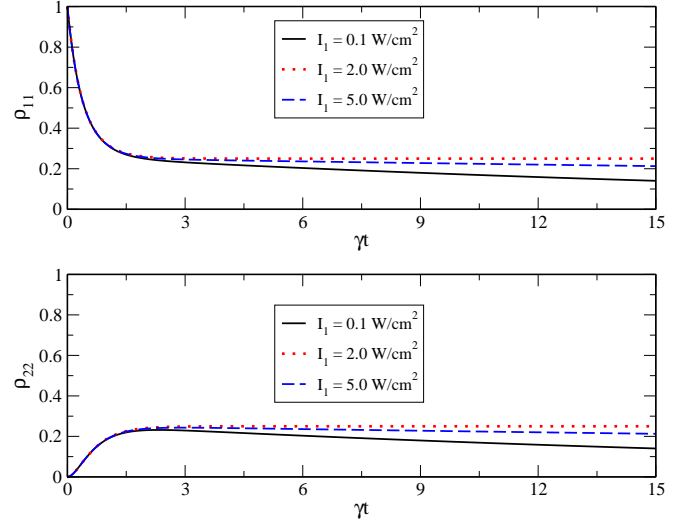


FIG. 3: (Color online) Same as in Fig. 2, but for different values of intensity of first laser, I_1 , keeping the intensity of second laser fixed at 0.1 mW cm^{-2} , keeping $\delta_1 = \delta_2 = 0$. The population is trapped at excited states for intensity of 2 W cm^{-2} .

trapping in the excited states. When $\delta_{12} = 0$, $\rho_{11}(t \rightarrow \infty) = \rho_{22}(t \rightarrow \infty) = [\rho_{11}(0) + \rho_{22}(0) - 2\text{Re}[\rho_{12}(0)]]/4$ become exactly same as normal VIC case [2]. It is worthwhile to emphasize that the results given in Eqs. (9), (10) and (12) are general because they are applicable to any PA coupling regime.

For experimental realization of VIC, our model can be applied to the spin forbidden intercombination transition $^1S_0 - ^3P_1$ of bosonic ^{174}Yb [24–26] which has no hyperfine interaction. The only molecular ground electronic state of ^{174}Yb is $^1\Sigma_g$ which corresponds to $^1S_0 + ^1S_0$ at long separation and represents the only bare continuum $|E\rangle_b$ of our model and . The excited states $|\phi_n\rangle$ can be chosen as ro-vibrational levels in long range O_u^+ state that can be populated by PA. For illustration, we specifically consider excited ro-vibrational levels $|\phi_1\rangle = |v = 118, J = 1\rangle$ and $|\phi_2\rangle = |v = 118, J = 3\rangle$ [25]. According to the selection rules of continuum-bound transitions, the minimum partial wave (l) that be coupled to $|\phi_2\rangle$ by PA is d wave ($l = 2$). Usually at ultracold temperatures, d wave scattering amplitude becomes insignificant due to large centrifugal barrier. But ground state scattering properties of ^{174}Yb are exceptional in the sense that it exhibits a prominent d -wave shape resonance at temperatures as low as $25 \mu\text{K}$ [24, 27].

We now discuss our numerical results. In Fig. 2, we show the dynamical behavior of populations ρ_{11} and ρ_{22} as a function of scaled time γt . The upper most panel shows populations in the absence of the lasers assuming $\gamma_1 = \gamma_2 = \gamma_{12} = \gamma = 2.29 \text{ MHz}$ [28]. The short time dynamics clearly shows exchange of population between $|\phi_1\rangle$ and $|\phi_2\rangle$ due to VIC. In the lower two panels, we plot ρ_{11} and ρ_{22} of Eq. (12) for different values of δ_2 , keeping δ_1 fixed. We find that, as δ_2 increases upto an optimum frequency, the lifetime of both the

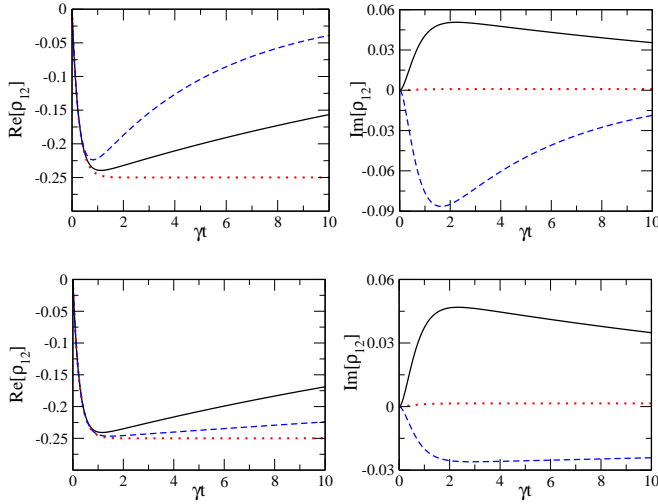


FIG. 4: (Color online) Plot of real and imaginary part of ρ_{12} against γt for different detunings of second laser (two upper subplots) and different intensities of the first laser (two lower subplots). All other parameters of the upper subplots is same as in Fig. 2 while the other parameters of lower subplots are the same as in Fig. 3. In the two upper subplots, the detuning $\delta_2 = 0$ (black solid line), $\delta_2 = 1$ MHz (red dotted line) and $\delta_2 = 3$ MHz (blue dashed lines) while in the lower two subplots the intensity $I_1 = 0.1 \text{ W cm}^{-2}$ (black solid line), $I_1 = 2.0 \text{ W cm}^{-2}$ (red dotted line) and $I_1 = 5.0 \text{ W cm}^{-2}$ (blue dashed lines)

excited levels also increases. Then at an optimum frequency, the population gets trapped in the excited state. For the parameters of Fig. 2, this optimum frequency is found to be 1 MHz. When δ_2 increases beyond the optimum value, population falls off. Since the value of dressed frequency $\tilde{\omega}_n$ depends upon the PA laser intensities, we expect the dynamics to be intensity dependent. Hence by varying the laser intensity of one of the PA lasers while keeping all other parameters fixed, we can achieve excited state population trapping for an optimum intensity of that laser. In Fig. 3, we show this explicitly for an optimized intensity $I_1 = 2 \text{ W cm}^{-2}$. Note that at this laser intensity, PA stimulated linewidth Γ_1 is much larger than γ_1 meaning that the system is in the strong-coupling regime. In Fig. 4, we plot the dynamical behavior of the coherence ρ_{12} as a function of γt . It is clearly visible that the imaginary part is much more smaller than the real part. The upper panel of Fig. 4 shows that $\text{Re}[\rho_{12}]$ becomes steady in the long time limit for an optimum frequency. Lower panel of Fig. 4 shows that $\text{Re}[\rho_{12}]$ becomes time-independent in the long time limit for the optimum parameters for which population in Fig. 3 becomes trapped.

In conclusion, we have demonstrated that it is possible to generate and manipulate coherence between two excited rovibrational states of a molecule by using the technique of PA spectroscopy. A promising candidate for exploring such excited state coherence is bosonic Yb atom. Once either or both the excited states are populated by PA, coherence between them builds up due to their interaction with the background

electromagnetic vacuum. We have analyzed the effects of PA lasers on VIC. Our results show that under certain conditions population can be trapped in excited states. Note that VIC can be best realizable for an idealized three level system. We have discussed VIC in an atom-molecule interface system where one of the levels is the collisional continuum of two ground state atoms. Our model relies on the condition that the two excited molecular levels decay to the same continuum. It may be very hard to fulfill this condition in alkali metal atoms since they have several ground continua due to hyperfine interactions. Furthermore excited levels may decay to bound states in ground molecular configuration. Since bosonic Yb has no hyperfine interaction, it has only one ground continuum. Photoassociated excited rotational levels with large vibrational numbers as considered in this work are unlikely to decay to any bound level since their Frank Condon overlap with the least bound state close to the ground continuum is very small. Therefore, bosonic Yb appears to be a promising candidate for exploring VIC. The manipulation of VIC with PA may be important for controlling decoherence and dissipation in cold molecules. It may also be used for coherent control of atom-molecule conversion and optical [29, 30] and magneto-optical Feshbach resonance [23].

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